Idaho Completion Project

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ICP/CON-04-00563
PREPRINT

High Hydrogen Concentrations Detected In The Underground Vaults For RH-TRU Waste At INEEL Compared With Calculated Values Using The INEEL-Developed Computer Code

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February 27 – March 3, 2005

Waste Management Symposium '05

HIGH HYDROGEN CONCENTRATIONS DETECTED IN THE UNDERGROUND VAULTS FOR RH-TRU WASTE AT INEEL COMPARED WITH CALCULATED VALUES USING THE INEEL-DEVELOPED COMPUTER CODE

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ABSTRACT

About 700 remote-handled transuranic (RH-TRU) waste drums are stored in about 144 underground vaults at the Intermediate-Level Transuranic Storage Facility at the Idaho National Environmental and Engineering Laboratory's (INEEL's) Radioactive Waste Management Complex (RWMC). These drums were shipped to the INEEL from 1976 through 1996.

During recent monitoring, concentrations of hydrogen were found to be in excess of lower explosive limits. The hydrogen concentration in one vault was detected to be as high as 18% (by volume). This condition required evaluation of the safety basis for the facility.

The INEEL has developed a computer program to estimate the hydrogen gas generation as a function of time and diffusion through a series of layers (volumes), with a maximum five layers plus a sink/environment. The program solves the first-order diffusion equations as a function of time. The current version of the code is more flexible in terms of user input. The program allows the user to estimate hydrogen concentrations in the different layers of a configuration and then change the configuration after a given time; e.g.; installation of a filter on an unvented drum or placed in a vault or in a shipping cask. The code has been used to predict vault concentrations and to identify potential problems during retrieval and aboveground storage. The code has generally predicted higher hydrogen concentrations than the measured values, particularly for the drums older than 20 year, which could be due to uncertainty and conservative assumptions in drum age, heat generation rate, hydrogen generation rate, G_{eff}, and diffusion rates through the layers.

INTRODUCTION

The Department of Energy (DOE) Carlsbad Field Office has set waste acceptance criteria, *Remote-Handled TRU Waste Acceptance Criteria for the Waste Isolation Pilot Plant* (WIPP), DOE/WIPP-Draft 18-3123, that must be satisfied before the INEEL RH-TRU waste drums can be disposed of at WIPP. One of the requirements for shipping drums from INEEL to WIPP is to ensure that hydrogen concentrations do not exceed 5% (by volume) in any volume of the waste drum during transport of the waste to the WIPP facility.

To address DOE Order 435.1 requirements, the INEEL developed a gas-generation computer model to calculate decay heat, hydrogen gas generation, and hydrogen concentration in a series of connected layers of packaging (volumes) in the waste drums as a function of time. Several sites have developed hydrogen generation models or performed analyses to suit their needs and waste streams. However, those analyses or models cannot be applied directly to the INEEL Argonne National Laboratory-East (ANL-E) waste stream. Computer code RadCalc 3.0 [1], which is used in transportation analysis for transport of waste in a CNS-10-160B cask [2], estimates hydrogen generation and concentration in a single volume without any leakage. This paper discusses the development of a computer code to estimate time-dependent hydrogen concentrations in a series of connected volumes. The current version of the code is an improvement over the previous version and has many additional features. This paper discusses the additional user-friendly features and compares the calculated and measured values of hydrogen concentration in the vaults.

The generation of hydrogen and mass transfer of hydrogen between layers/volumes (maximum five plus sink) of the confinement is modeled. The model assumes that all hydrogen gas generation occurs within the innermost layer of the confinement, i.e., layer 1. The accumulation rate of hydrogen within the innermost layer of confinement is defined by the hydrogen generation rate and gas transport rate from that volume.

RH-TRU WASTE

Cleanup operations were conducted after a certain number of fuel element cutting and grinding operations were performed in the hot cell (at least annually, or as required by facility workload). Waste items were placed in containers loose, plastic-bagged, containerized 4-in. (10.2 cm)-diameter by 10-in. (25.4 cm)-high portal cans, or in unsealed 4-L paint cans. When a waste collection can was filled, it was moved to another workstation within the hot cell, where it remained until sorted and fuel/cladding or other high-dose rate pieces were removed. ANL-E

personnel also visually verified that no prohibited items were present, such as compressed gases and liquids. In packaging RH-TRU waste material at ANL-E [3], items were first segregated as combustible (C) or noncombustible waste (NC). If any high-activity material (>30 rem, R/hr (0.2 Sv/hr)) was present, it was removed and disposed of with the recoverable scrap or swarf.
Some metal and glass containers were compacted. The glass was placed inside a metal container (portal can) before compaction. The ratio of volume reduction ranged from 4:1 to 8:1. The combustible waste was not compacted. The packaging materials, including two 7.5-gallon (28-L) containers and the 30-gallon (114-L) steel drum, weigh about 64 lb (29 kg).
Of the 700 RH-TRU waste drums, 617 drums came from ANL-E. The waste was generated by destructive examinations of irradiated fuel elements at the Alpha-Gamma Hot Cell Facility. These drums are stored in about 144 underground vaults of various sizes, ranging from 4- to 11-drum capacity.

WASTE PACKAGING CONFIGURATION

Briefly, a typical ANL-E waste packaging was configured as follows (Figure 1):

Waste is placed into two 28-L waste cans, without a gasket in the can lid. The cans were equipped with toggle-latch-cover clamp rings.
The 28-L cans are placed into a 90-mil (2.3-mm) lidless fiberboard liner.
The liner and cans are placed inside a 20-mil (0.5-mm) polyvinylchloride (PVC) bag that is heat-sealed closed, although a 60-mil (1.5-mm)-thick polyethylene disc puncture guard was placed over the PVC pouch (innermost volume).
This package is placed into a 100-mil (2.5-mm) polyethylene (PE) lidless liner.
The PE liner and contents are placed inside a 20-mil (0.5-mm) PVC bag that is heat-sealed closed (middle volume)
This package is placed into a 114-L drum (DOT 7A, Type A). After a lid was placed on a container, it was surveyed. Any container with a reading greater than 30 R/h (0.3 Sv/hr) was repackaged. The drum lid-gasket is 3/8-in. (9.5-mm) OD × 7/32-in. (5.6-mm) ID styrene-butadiene rubber.

Waste drums prepared before 1989 typically contained heat-sealed bags that did not have a filter vent, whereas drums prepared after 1989 had bags with a filter vent and a twist-and-tape or fold-and-tape closure [3]. Some drums may not have a filter vent in the drum lid. After July 1983, the PVC bags were lined with Kraft paper liners.
About 187 of the 617 114-L drums (DOT 7A, Type A) are vented with an NF-013 carbon composite drum filter in the drum lid (not shown in the Figure 1). Of these 187 drums, about 121 contained PVC bags vented with an NF-030 carbon composite filter (not shown in the Figure 1), and 66 contained heat-sealed bags with no filters. The remaining 430 drums contained no filters in the bag or in the drum lid.

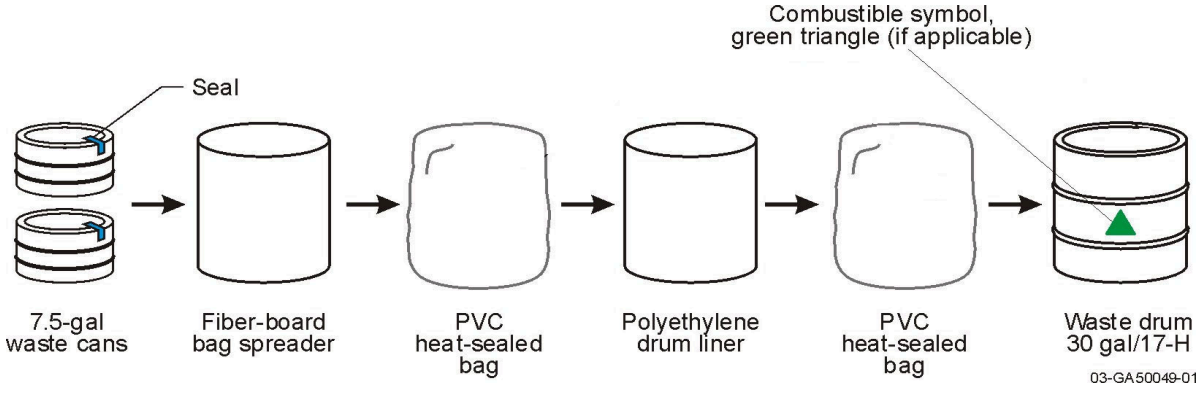

Figure 1. Schematic of 30-gallon (114-L) drum packaging.
Hydrogen transport across the layers and hydrogen generation rate processes are defined by a series of system equations, as discussed in the following two sections.

HYDROGEN TRANSPORT PROCESS

The generation of hydrogen and mass transfer of hydrogen between layers (maximum five layers plus sink) of confinement is modeled. It is assumed that all hydrogen gas generation occurs within the innermost layer of the

confinement, i.e., layer 1. The accumulation rate of hydrogen within the innermost layer of confinement is defined by the hydrogen generation rate and gas transport rate from that volume:

$$\frac{dn_{H_2,1}}{dt} = n_{H_2,gen} - L_{R,1\to 2}\left(\frac{n_{H_2,1}}{n_{T.1}} - \frac{n_{H_2,2}}{n_{T,2}}\right) \quad (1)$$

where

$n_{H_2,j}$ = moles of hydrogen within the jth void volume

$n_{H_2,gen}$ = hydrogen generation rate, mol s^{-1}

$L_{R,1\to 2}$ = hydrogen gas transport rate across from layer 1 to 2 of drum, mol s^{-1} (mol fraction)-1

$n_{T,j}$ = moles of total gas in j^{th} void volume

t = time, s.

Hydrogen gas accumulation within other layers of a drum can be defined by a generic equation, as follows:

$$\frac{dn_{H_2,j}}{dt} = L_{R,j-1\to j}\left(\frac{n_{H_2,j-1}}{n_{T.j-1}} - \frac{n_{H_2,j}}{n_{T,j}}\right) - L_{R,j\to j+1}\left(\frac{n_{H_2,j}}{n_{T,j}} - \frac{n_{H_2,j+1}}{n_{T,j+1}}\right) \quad (2)$$

where $n_{H_2,j}$ and $n_{T,j}$ are the moles of hydrogen and total gas within the j^{th} layer of confinement, respectively, and j is equal to 2 to 5. The moles of hydrogen in the sink volume are assumed to equal zero at all times.

The user provides the starting time, end time, time step (dt), and number of time steps to print the data. The volume (cm^3), hydrogen concentration (%) in each volume, pressure (psia), and temperature (^oF) are user input. Based on these parameters, the code initializes the number of hydrogen and air moles using Ideal Law in each volume. The subroutine NH3 calculates diffusion in layers.

The FORTRAN computer code was developed to solve the system of differential equations using the Runge-Kutta-Fehlberg numerical method [4].

HYDROGEN GENERATION RATE

Hydrogen is generated primarily via the radiolysis of hydrogenous materials in the waste. In the *Safety Analysis Report for the RH-TRU 72-B Waste Shipping Package* (72-B Shipping Package SAR) [5], an equation defines the rate of hydrogen generation as:

$$n_{H_2,gen} = C\sum_i [F_i G_i (DH)_i] \quad (3)$$

where

C = conversion constant = 1.04×10^{-7} (g-mol) (100 eV)/(molecule)(W-s)

F_i = fraction of emitted energy from type i radiation absorbed in waste material

G_i = number of molecules of hydrogen produced per 100 eV of energy absorbed from type i radiation

DH_i = decay heat (W) of type i radiation.

The methodologies for determining the value of each parameter in Equation (1) are described below.

FRACTION OF EMITTED ENERGY ABSORBED IN THE WASTE MATERIAL

The fraction of emitted energy absorbed in the waste material for type i radiation, F_i, is a function of the fraction of emitted energy that escapes the radioactive particles, f_i, and the fraction of escaped energy absorbed by the waste material, ϕ_i:

$$F_i = f_i \phi_i \quad . \qquad (4)$$

In the case of alpha radiation, as a result of self-shielding a fraction of the alpha radiation is attenuated. The fraction of emitted energy that escapes the radioactive particle, f_α, is a user-defined parameter. Most beta radiation will escape the radioactive waste material and is not subject to self-shielding. Therefore, the fraction of emitted energy that escapes the radioactive material, f_β, is assumed to be unity. However, all escaped alpha (Φ_α) and beta (Φ_β) energy is assumed be absorbed in the waste material because of its mass and volume and is assumed to be unity. Thus, the fraction of emitted α-energy, F_α, and β-energy, F_β, absorbed in waste material, is equal to f_α and 1, respectively.

Determining the gamma-absorbed dose is more complex than for alpha and beta radiation, because the total energy of the gamma emission might not be deposited in the waste form. In the case of gamma radiation, no radiation is attenuated by the radioactive particles, and therefore the fraction of emitted energy that escapes the radioactive particles, f_γ, is unity. Thus, F_γ equals ϕ_γ.

Isotopic distributions obtained from the ORIGEN2 [6] analysis indicate that at the time of drum loading, 1 year after the irradiation, about 10% of gamma radiation is emitted from low-energy isotopes (<0.1 MeV) and 90% from higher-energy isotopes (>0.1 MeV). High-energy gamma radiation is primarily emitted from two isotopes, each with about 0.5 MeV in energy. It was also observed from ORIGEN2 calculations that gamma radiation emitted by low-energy isotopes decreases as a function of time, t, until after 5 years most radiation is emitted by higher-energy isotopes. The total fraction of gamma radiation absorbed as a function of time, $\phi_{\gamma,t}$, is calculated by the following equations:

$$\phi_{\gamma,t} = [0.9 + (\frac{t}{5})0.1]\phi_{0.5MeV} + 0.1[1 - (\frac{t}{5})]\phi_{0.1MeV}$$

; t < 5 yr (5)

$$\phi_{\gamma,t} = \phi_{0.5MeV}$$

; t ≥ 5 yr . (6)

The fraction of gamma radiation emitted by low-energy radioisotopes absorbed by the waste, $\phi_{0.1MeV}$

, is assumed to equal unity. Therefore, F_γ for low energy gamma is assigned to be unity.

A method for calculating the absorbed gamma dose associated with 46 radionuclides commonly used in radioactive waste analysis and transport has been developed that accounts for the physical characteristics of the waste and the waste container geometry [7]. Gamma energy absorbed by the waste by each nuclide is a function of the abundance of gamma rays per decay for each nuclide, the energy associated with each gamma ray, and the fraction of energy from each gamma ray that is absorbed in the waste. A previously developed model calculates the fraction of gamma radiation absorbed in the waste material, ϕ_γ, as a function of the gamma radiation energy, the characteristic packaging dimension, and the waste density [7]. The polynomial equation to calculate the fractional gamma energy absorption of each nuclide, shown in Equation (7), was developed, which is derived from statistical-analysis-predicted gamma absorption in hypothetical containers with varying geometries. We did a nonlinear regression analysis of 80 data points to determine the equation coefficients over a range of gamma radiation energy from 0 to 2 MeV, for a characteristic packaging dimension between 0.29 and 0.91 M, and waste densities between 0.6 and 2.0 g cm^{-3}:

$$\phi_\gamma = 1 + [A_1E + A_2E^2 + A_3E^3][\frac{A_4}{L} + \frac{A_5}{L^2} + \frac{A_6}{L^3}][\frac{A_7}{\rho} + \frac{A_8}{\rho^2} + \frac{A_9}{\rho^3}] \quad (7)$$

where A_1 through A_9 are constants, E is gamma radiation energy (0.5 MeV), L is the characteristic packaging dimension in feet, and ρ, is the waste density in g cm^{-3}.

The characteristic packaging dimension, L, and the waste density, ρ, are user-defined parameters. Conservatively, a radius or an equivalent radius of packaging is used as the characteristic packaging dimension. The subroutine GAMMAABS calculates the fraction of gamma radiation emitted by high-energy radioisotopes, $\Phi_{0.5MeV}$, absorbed by the waste as a function of the characteristic packaging dimension (L, ft) and the waste density (ρ, g cm^{-3}).

Note that all the hydrogen generated from gamma radiation is assumed to be in the innermost volume.

DECAY HEAT GENERATION RATE IN THE WASTE

The heat generation rate in the waste is a function of the mass of the radioisotopic inventory. Normally, the RH-TRU radioactive waste constituents are the long-lived actinides and fission products. The waste is generated as result of cutting and grinding irradiated fuels/materials. For development of the heat generation model, we selected the waste in ten drums, ANL-728 through -737 [8]. Total radio-isotopic distributions in each pin/material at the time of drum loading were provided by the waste generator. We estimated the total mass from each pin/irradiated material that went into the waste drums based on the number of cutting and grindings [9,10]. Most of the decay heat from alpha radiation is produced from the decay of actinides, which are relatively long-lived compared to fission products. Most decay heat from the beta and gamma radiation is produced from the decay of fission products, which are relatively short-lived compared to actinides. Since the actinides are relatively long-lived compared to the period of interest (50 years), the inventory of actinides can be estimated based on the original inventory and burnup of fissile material. Fission products produced is directly proportional to the amount of fissile material that undergoes fission and decay time. Three decay heat algorithms were developed, one for the heat generation from alpha radiation and one each for the heat generation from the beta and gamma radiation, as shown in Equations (8), (9), and (10). In developing these models, we considered a total of 26 fission and activation products and eight actinides. Only those isotopes that contributed more than 0.1% of the total heat generation rate at any time during the 50-yr period after the drum loading are included.

$$DH_\alpha = Pu-239 \times [2.114 \times 10^{-7} t^3 - 2.26 \times 10^{-5} t^2 + 1.011 \times 10^{-3} t + 5.346 \times 10^{-2}] \quad (8)$$

$$DH_\beta = MFP \times [2.17 \times 10^{-3} e^{-0.7805t} + 5.136 e^{-0.02366t}]$$

$$DH_{\gamma} = MFP \times [4.68 \times 10^{-4} e^{-0.697t} + 2.66 \times 10^{-4} e^{-0.0231t}] \quad (9)$$

(10)

where

DH_{α}, DH_{β}, and DH_{γ} = decay heat generation rate from alpha, beta, and gamma radiations, respectively, in W

Pu-239 = inventory of PU-239 at the time of drum loading, Ci

MFP = fission products at the time of drum loading, Ci.

Some radioisotopes have more than one decay mode. Table I lists the radioisotopes included in the analysis and their decay modes. The alpha, beta, and gamma decay heat rate fractions for individual isotope were obtained from Reference 11.

Table II lists the heat generation rates as a function of time. Decay heat generation rates from different radiation modes in a typical ANL-E RH-TRU waste drum are plotted in Figure 2.

The subroutine DEKHEAT calculates the decay heat rates from alpha, beta, and gamma radiation.

HYDROGEN GAS GENERATION RATES

In the case of a waste containing M types of hydrogenous materials, an effective hydrogen gas generation rate Geff-value is defined in terms of the waste-specific G-values [5]:

$$G_{eff,i} = \sum_{m}^{M} \bar{f}_m G_{m,i} \quad (11)$$

where $\bar{f}_m$ is the fraction of energy from radiation type i absorbed by material m, and $G_{m,i}$ is the hydrogen generation value for radiation type i in material m.

Many models simulating hydrogen generation conservatively assume constant G-values. The G-values of various hydrogenous materials have been tabulated in the *Safety Analysis Report for the RH-TRU 72-B Waste Shipping Package* [5].

The maximum G-values (independent of radiation type) for polyethylene and dry cellulose are 4.1 and 3.2 molecules/100 eV, respectively. Radiolysis experimental results indicate that the hydrogen-generation potential of many organic materials decreases as a function of total dose, D(t), due to waste matrix depletion [12]. Waste matrix depletion describes the decrease in hydrogen gas generation potential of material as hydrogen in the hydrogenous material is depleted by radiolytic reactions.

Table I. Actinides, fission, and activation products associated decay modes in the waste*

Mn-54	(γ)	Sb-125	(β, γ)	Eu-155	(β, γ)
Fe-55	(β, γ)	Te-125m	(β, γ)	U-234	(α)
Co-60	(β, γ)	Te-127	(β, γ)	U-235	(α, β, γ)
Sr-89	(β)	Cs-134	(β, γ)	U-238	(α, β, γ)
Sr-90	(β)	Cs-137	(β)	Pu-238	(α)
Y-90	(β)	Ba-137m	(β, γ)	Pu-239	(α)
Y-91	(β)	Ce-144	(β, γ)	Pu-240	(α)

Zr-95 (β, γ)	Pr-144 (β, γ)	Pu-241 (β)
Nb-95 (β, γ)	Pr-144m (β, γ)	Am-241 (α)
Ru-106 (β)	Pm-147 (β)	Cm-242 (α)
Rh-106 (β, γ)	Sm-151 (β)	Cm-244 (α)
Sn-123 (β, γ)	Eu-154 (β, γ)	

*A contribution of less than 0.05% within an isotope is not included.

Table II. Heat generation rates in ANL-E RH-TRU waste stored at INEEL.

Time (y)	Alpha (W) /Ci Pu-239	Beta (W) /Ci-MFP	Gamma (W) /Ci-MFP
	Equation 7	Equation 8	Equation 9
0	5.346E-02	2.684E-03	7.34E-04
1	5.445E-02	1.496E-03	4.93E-04
2	5.539E-02	9.454E-04	3.70E-04
5	5.798E-02	5.001E-04	2.51E-04
10	6.152E-02	4.063E-04	2.12E-04
15	6.424E-02	3.602E-04	1.88E-04
20	6.631E-02	3.200E-04	1.68E-04
25	6.788E-02	2.843E-04	1.49E-04
30	6.911E-02	2.526E-04	1.33E-04

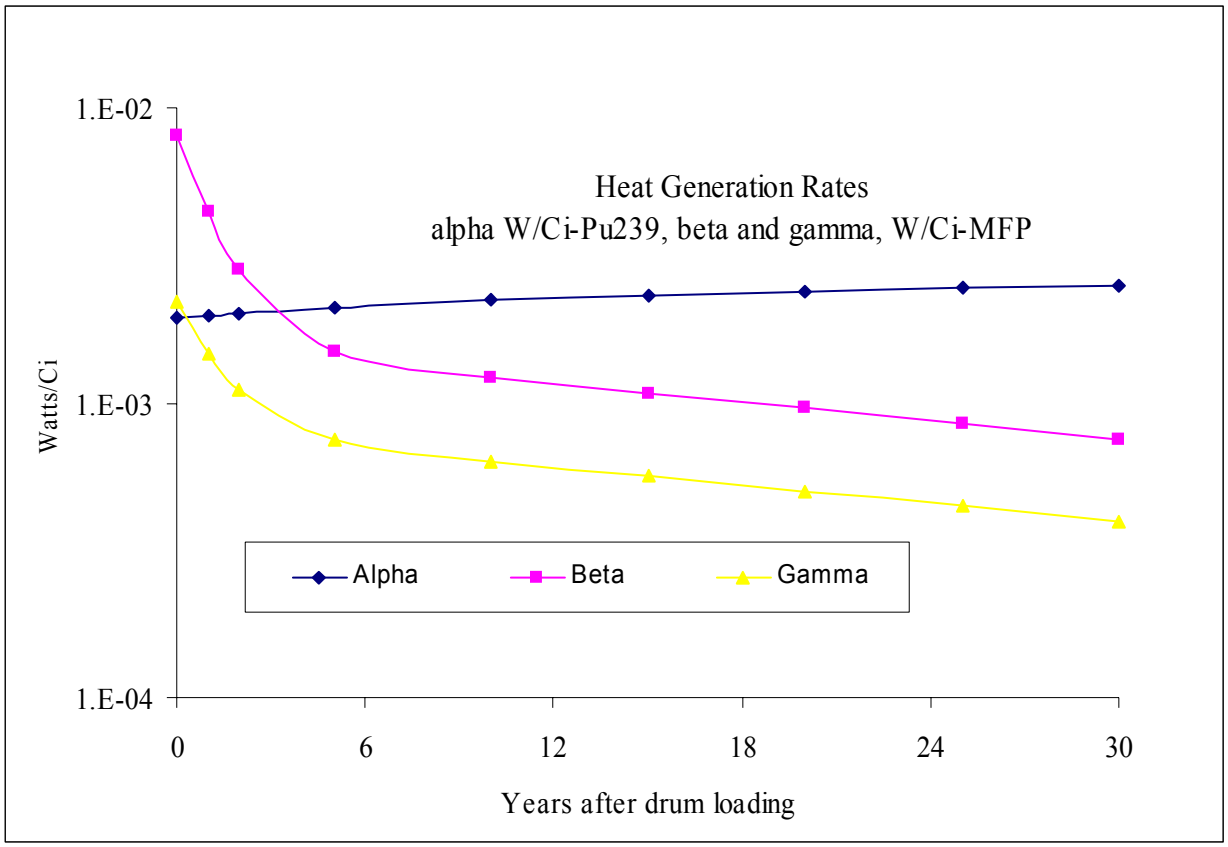

Figure 2. Decay heat generation rates in a typical RH-TRU waste drum stored at INEEL.

We developed an expression for G-value, molecules/100 eV, for continuous matrix depletion using experimental data from matrix depletion studies [12].

$$G = k_1 . e^{-k_2 D(t)} \qquad (12)$$

where k_1(molecules/100 eV) and k_2(W-yr)$^{-1}$ are matrix-dependent constants, and D(t) is total dose in W-yr. For polyethylene, the constants k_1 and k_2 are derived to be 2.34 and 1.45, and for dry cellulose 0.934 and 143.1, respectively, from the INEEL matrix depletion study [12].

G-values for α, β, and γ-radiation are user-defined inputs, each defined by two parameters. In the case of the matrix depletion model (step function model), the first parameter constant (k_1) defines the nonmatrix depletion value, and the second constant (k_2) represents the matrix depleted value. The code uses the matrix depletion values after a total dose of 0.012 W-yr. The user can model more than one source in the waste drum; e.g., two 28-L buckets in the ANL RH-TRU waste drums. In this case, the code will use the matrix depletion value to be 0.024 W-yr (2×0.012). For a constant G-value, a constant k_2 will be zero.

The subroutine EFFGVALU calculates the hydrogen generation rate G_{eff} for alpha, beta, and gamma radiations. Substituting Equations (4), (8), (9), (10), and (11) into Equation (2) and simplifying the resulting equation yields an expression describing the hydrogen generation rate, $n_{H_2,gen}$:

$$n_{H_2 gen} = C[f_\alpha G_{eff,\alpha}(DH)_\alpha + G_{eff,\beta}(DH)_\beta + \phi_\gamma G_{eff,\gamma}(DH)_\gamma$$

. (13)

The subroutine NH2 calculates the total hydrogen generation rate in the waste.

HYDROGEN TRANSPORT RATE

The hydrogen gas transport rate, L_R, from layer j to j+1 (mol s^{-1}/mol fraction^{-1}) is user-provided input. In the case of a vented layer, it would be the diffusion rate of the filter. For an unvented layer, the hydrogen gas transport rate,L_R, can be estimated using the following equation:

$$L_{R,P} = \frac{\wp A_p P c}{x_p} \quad (14)$$

where

$\wp$ = hydrogen gas permeability, cm^3(STP) cm^{-1} s^{-1} (cm Hg)$^{-1}$

A_p = layer (bag) permeable area, cm^2

P = gas pressure, cm Hg

c = gas concentration, mol cm^{-3}

x_p = layer (bag) thickness, cm.

This computer code was used to estimate hydrogen concentration in the ANL-E RH-TRU waste drums.

INPUT PARAMETERS AND ASSUMPTIONS

The drum-packaging scheme shown in Figure 1 was used in a model to calculate hydrogen generation in unvented DOT 17-H 30-gallon (114-L) waste drums. In the case of vented drums and vented bags, filter diffusivities were used.

A number of assumptions were made in estimating absorbed gamma radiation. Since the characteristic packaging dimension (drum radius) of a 114-L drum is less than the lower limit values used to define the coefficients, conservatively, the lower limit (L = 0.29 M, the same as a 55-gal (208-L) drum is used. Liekhus [13] estimated the waste densities to be between 0.26 and 0.73 g cm^{-3}, using the weight range for 114-L drums and the drum liner volume containing waste described by INEEL content code IDC 104. The maximum waste density of 0.73 g cm^{-3} is used in all calculations.

The total volume of the innermost layer (inner heat-sealed bag) is estimated to be 98.4 L [14]. The innermost volume contains two 28-L pails of waste material. The total volume occupied by the waste and drum liner is assigned to be 28.4-L. Therefore, the void volume in the innermost layer would be 70 L. The remaining two layers [middle layer (between the two plastic bags) and outer layer (between the second plastic bag and drum)] are assigned a void volume of 7.6 L each. The middle layer is the void volume between the two polymer bags. The outer layer is the void volume between the drum and the polymer bag.

The quantity of waste composition is often unknown; therefore, it is assumed that all organic waste consists of polyethylene. Polyethylene has the highest hydrogen-generating potential of any material expected in the waste drum. The G-value, described in Reference 5, is a function of the total number of molecules produced for 100 eV, dose, and radiation type.

In the case of combustible material in the drum for alpha and beta radiation, the maximum non-matrix-depleted G-value is assumed to be 4.1 molecules/100 eV up to the total dose of 0.012 W-yr [15]. After the matrix depletion, the effective G-value of 1.09 molecules/100 eV, constant for alpha and beta radiation, is used. This value is the highest G-value associated with matrix-depleted organic material (i.e., wet cellulose). The gamma radiation G-value

is assumed to remain constant and be independent of the total radiation dose. This assumption is based on the large mass of polymeric packaging material, as well as on the significant decrease in the fraction of absorbed gamma radiation with increasing gamma radiation energy. For gamma radiation, a G-value of 4.1 molecules/100 eV, constant, is assigned. Note that all the hydrogen generated from gamma radiation is assumed to be in the innermost volume.

In the case of noncombustible material, it is conservatively assumed that at least 20% of the waste is combustible. Therefore, 20% of the G-values were used for alpha, beta, and gamma radiation [16].

The hydrogen leak rate across the heat-sealed polymer bags, $L_{R,P}$, is a function of hydrogen gas permeability across the polymer, bag thickness, bag surface area, and gas concentration [17]. The polymer bag thickness is 5×10^{-2} cm [12]. Hydrogen gas permeability across this polyethylene bag is 7.6×10^{-10} (cm^3 (STP) cm^{-1} s^{-1} kPa^{-1}) [18]. The surface area of a polymer bag is estimated, based on the cardboard liner diameter of 42 cm and height of 100 cm, to be 1.5×10^4 cm^2. Assuming standard temperature and pressure (STP), pressure and gas concentrations are 101 kPa, and 4.1×10^{-5} moles/cm^3, respectively. It is conservatively assumed that there is no significant pressure buildup inside the bags. In the case of bags taped closed, hydrogen leak rates are on the order of 10^{-6} to 10^{-7} mol/s/mol fraction. For the purpose of this analysis, the leak rate across polymer bags of 2.32×10^{-7} mol/s/mol fraction was used [19]. As discussed, some bags were vented, and a leak rate of 3.7×10^{-6} mol/s/mol fraction was assigned [20].

Measured hydrogen diffusion characteristics across unvented 208-L drum lid gaskets were 10^{-6} to 10^{-7} mol/s/mol fraction [21]. It is assumed that the leak rate across the lid gasket is proportional to the circumference of the gasket; thus, the hydrogen diffusion characteristic across the gasket of a 114-L drum is 10^{-7} mol/s/mol fraction. This leak rate was used for all the unvented drums. As discussed, some drums were vented, and a leak rate of a 3.7×10^{-6} mol/s/mol fraction was assigned.

For this exercise, the estimated radionuclide inventory of an individual drum was based on the batch average inventory and drum surface dose rate reported at the time of shipping. Appendix D of the AK documentation report for INEEL RH TRU waste from ANL-E [12] lists the consolidation batch, number of drums in that batch, average radioisotope inventory (mainly Pu-239 and U-235), waste type, drum packaging date, and drum-specific dose rate, etc.

First, the data were sorted on the batch basis. Next, average dose rates were calculated. Based on average inventory and the ratio of drum dose rate to average dose rate, Pu-239 and U-235 inventories in individual drums were then estimated. The mixed fission product activity is estimated using the algorithm developed in Reference 20, which is based on ANL-E RH-TRU waste drums 728–737 inventories [16]. A ratio of mixed fission products to grams of fissile material (Pu-239 + U-235) in the waste was estimated to be 2.55 Ci of MFP. Similarly, based on the drum packaging date, the drum age as of October 31, 2004 was estimated. All unvented drums were more than 16 years old.

We made the following assumptions in estimating the hydrogen concentration in four-, five-, and eleven-drum vaults:

A one-year decay period is assumed before the waste was placed in the drums that are more than 16 yrs old.

For the drums shipped in the late 1980s and 1990s (15 years old), ANL-E procedure used 6.5 years of decay to estimate fission product for the shipping documents. Therefore, for our analysis we used 6 years of decay period before the waste was placed in drums. [22].

The lag time between the drum packaging and storage in the vault is one year.

The vault void volume is simulated based on the maximum number of drums in a vault. For example, for a four-drum vault the minimum vault void volume is estimated to be 81,656 cm^3 per drum. (The vault dimensions are 10.5 ft (3.2 M) high and 2 ft (0.61 M) in diameter, and a drum's dimensions are 29.5 in. (0.75 M) high and 20 in. (0.51) in diameter.)

The total vault leakage rate was determined based on initial measurements on a few vaults. Several runs were made using different leakage rates until the predicted hydrogen concentration in the vault matched the measured value. Based on the comparison, total leakage rate from the vault was estimated to be 2.5×10^{-8} mol/s/mol fraction.

We assumed all the drums were placed in a particular vault at the same time. The history of vault operations is unknown. We assumed no vaults were opened after they were closed.

First, the average radioactive material inventory in a drum was estimated, based on the total number of drums and the inventories in individual drums. Then, the hydrogen concentration in a drum was calculated, assuming the drum

was stored above ground for 1 year before being placed in the vault. Next, the run was made using the hydrogen concentrations in the three layers of a drum at the time of vault loading and drum storage time in the vault. Similarly, the process was repeated for different vaults. Tables III and IV list sample input and output files.

Table III. Sample input file.

'Vault 51A - 5 drums Combust/noncomb weighted inv @ ~1yr'	
'A51Ph1.out'	!Output File Name
0,1,1000,0.0001	!Elapsed time after drum loading, End time, Print step, Time Step
3,70030.,7571.,7571.	!Number of volume (nvol), Volnme(i=1,nvol), cm^3
1,0,0,	!Volume # for Max H2, H2 Concentration(i=1,nvol)
0,9.956e-4,.2605,2	!Decay heat f(t), Initial Ci loading (Pu-239, MFP), # of sources
0.73,0.95,12.5,65	!Waste Density (g/cm^3), Pack. Parameter, press (psi), Temp (F)
2.32e-7,2.32e-7,3.7e-6	!Leak rate(i=1,nvol)
1,.82,1	!G-step function, Alpha Escape fraction, Gamma model
3.4,1.09,3.4,1.09,4.1,4.1	!G-values for alpha, beta, and gamma

CONCLUSIONS

Comparison of Measured Values with the Code-Predicted Values

The code consistently predicted higher values than those actually measured. Table V lists the measured values versus the code-predicted values, which are plotted in Figure 3. Considering the significant uncertainty in drum age, particularly for the drums older than 20 years, the results for inventories, leakage rates, hydrogen generation rates, and measurements are in good agreement, except for the older drums. The results should be interpreted as approximations rather than absolute values. Since the analyses were performed as the safety analyses for the operation of vaults, and due to lack of sufficient information, most parameters used in the analysis are conservative. The code was used in designing an intermediate storage container for RH-TRU waste drums that were retrieved from the vaults and stored above ground [20]. The code was also used in estimating the time hydrogen concentration would built to 2.2% in sealed ILTSF vaults after purging, so the monitoring frequency could be established [19].

Comparison of the INEEL Code with Radcalc 3.0

RadCalc [1] is a computer code widely used in transportation analysis by the Department of Energy and many of its sites. The code simulates a single volume only; it does not simulate the leakage from that volume. The inventory of individual isotopes in the waste is an input requirement. RadCalc has built-in libraries for decay constants, decay heat rates for radioactive isotopes, and the hydrogen generation rate from individual waste material types; e.g., alcohol, cement, etc. RadCalc users have the choice of either inputing the values of hydrogen generation rates for each radiation type or selecting the material type, and

Table IV. Sample output file.

```
Radwaste Hydrogen Generation, Diffusion, and Concentration Analysis  08-OCT-04

Vault 51A - 5 drums Combust/noncomb weighted inv @ ~1yr

Input file Name = a51ph1.txt

              Waste Packaging Parameters
 Volume 1     Volume 2     Volume 3     Volume 4     Volume 5
  70030.0      7571.0       7571.0
        Initial Hydrogen Concentration (%)
     0.0          0.0          0.0
Waste Density (gm/cm^3) = 0.730  Crit. Packg. Dimension, ft =  0.95
Pressure (psia) = 12.500  Temperature (F) = 65.000

                Leak rates  (mol/s/mol fraction)
      Vol1-2     vol2-3      Vol3-4       Vol4-5      Vol5-
     2.32E-07    2.32E-07    3.70E-06

              Decay Heat Parameters
Decay heat curves are used.
Pu-239, Ci =  0.001  MFP, Ci =  0.261  Number of Waste Packages =  2
```

Table V. Comparison between predicted and measured values.

Vault Number	Number of Drums	Average Age of Drum	Average Pu-239 (Ci)	Average MFP (Ci)	Waste Type*	Hydrogen Concentration (%)	
						Predicted	Measured
A51	5	19	1.90E-03	0.858	C/NC	0.6	0.8
B49	5	19	1.71E-03	0.544	C	1.2	1.3
A17	10	26	1.29E-02	2.09	C	1.6	2.7
E6	4	9	9.22E-03	1.33	C/NC	2	2.7
A53	5	14	1.26E-02	1.68	C	3.6	1.2
A55	5	13	1.61E-02	1.89	C	3.8	3.3
F8	4	9	2.85E-02	4.22	C	4.1	1.3
B21	4	24	2.11E-02	2.62	C	4.6	5.2
B52	5	14	1.27E-02	1.7	C	5.6	2
C16	11	24	0.144	18.8	NC	6.4	0.8
D41	5	20	4.27E-02	5.65	C	8.3	4.8
E42	5	21	8.00E-02	7.35	C	11.6	4.4

C = combustible; NC = noncombustible.

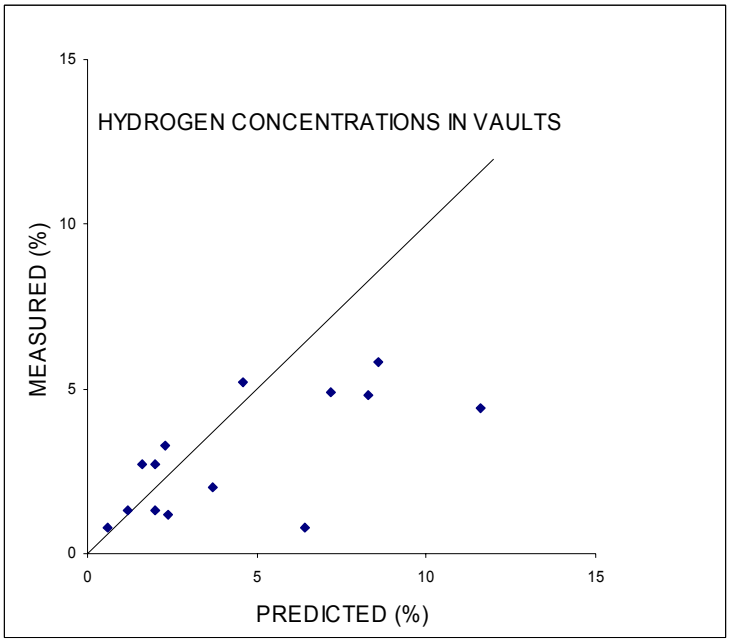

Figure 3. Comparison of the code-predicted and measured values of hydrogen concentration.

RadCalc will calculate the hydrogen generation rate. RadCalc outputs only the rates for an initial and final time. Its output is not a function of time. We repeated the RadCalc calculation 20 times to obtain the values presented in Table VI. The INEEL-developed code outputs all of this information as a function of time.

Based on our model and RadCalc, we made the following comparisons for a single volume and no leakage. As seen from Table VI, the INEEL's model predicted values match within 1% of the values

predicted by RadCalc.

Table VI. Comparison of INEEL's model with RadCalc .

Time (y)	Heat Generation Rate (W) × 10^{-2}		Hydrogen Generation Rate (Cm^3/hr)		Hydrogen Concentration (% Volume)	
	INEEL	RadCalc	INEEL	RadCalc	INEEL	RadCalc
0	3.64	3.65	0.333	0.332	0.	0.
1	2.37	2.34	0.216	0.213	3.23	3.2
2	1.78	1.77	0.162	0.162	5.36	5.3
3	1.5	1.51	0.136	0.134	6.98	6.9
4	1.37	1.39	0.124	0.126	8.37	8.3
5	1.30	1.32	0.119	0.120	9.62	9.6
6	1.27	1.29	0.116	0.117	10.8	11
7	1.25	1.27	0.114	0.115	11.93	12
8	1.24	1.25	0.113	0.114	13.02	13
9	1.23	1.24	0.113	0.113	14.08	14
10	1.22	1.23	0.112	0.112	15.1	15

REFERENCES

Duratek Federal Services, *Radcalc 3.0*, DFSNW-RPT-042, Vol. 1, Rev. 0, Richland, Washington, November 2001.

GTS Duratek, *Safety Analysis Report for Chem-Nuclear Systems Model CNS 10-160B Type B Radwaste Shipping Cask*, Rev. 18, Columbia, South Carolina, June 2002.

INEEL, *AK Documentation Report for INEEL-Stored Remote-Handled Transuranic Waste from Argonne National Laboratory-East*, Rev. 0 (Draft Final), prepared by IT Corporation, Albuquerque, New Mexico, July 15, 2003.

M. E. Davis, *Numerical Methods and Modeling for Chemical Engineers*, New York: John Wiley & Sons, 1984.

Westinghouse, *Safety Analysis Report for the RH-TRU 72-B Waste Shipping Package,* Rev. 1, Westinghouse Electric Corporation, Waste Isolation Division, Carlsbad, New Mexico, August 1999.

G. Croff, *ORIGEN2 - A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code*, ORNL-5621, Oak Ridge National Laboratory, Oak Ridge, Tennessee, July 1980.

EPRI, *Methodology for Calculating Combustible Gas Concentration in Radwaste Containers*, EPRI NP-4938, Electric Power Research Institute, Palo Alto, California, March 1987.

INEEL, *Radiological Acceptable Knowledge Report for Drums 728–737*, INEEL/EXT-00527, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho, July 2002.

S. T. Khericha and P. Kuan, *Results of ORIGEN2 Calculations for 14 EBR-II Experiment Fuel Elements that Generated Remote-Handled TRU Waste in Drums 728–737*, Idaho National Engineering and Environmental Laboratory, Engineering Design File-2365, September 2002.

S. T. Khericha and P. Kuan, *Estimation of Inventory of Significant Isotopes in RH-TRU Waste Drums 728 through 737*, Idaho National Engineering and Environmental Laboratory, Engineering Design File 1979, June 2002.

DOE, *Integrated Data Base Report-1993: U. S. Spent Nuclear Fuel and Radioactive Waste Inventories, Projections, and Characteristics*, DOE/RW-0006, Rev. 1, Department of Energy, 1994.

INEEL, *TRUPACT-II Matrix Depletion Program Final Report*, INEEL/EXT-98-00987, Rev. 1, Lockheed Martin Idaho, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho, October 1999.

K. J. Liekhus, correspondence with M. E. McIlwain, "Results of Experiments at Argonne to Determine Effective G-Value from RH-TRU Waste Drums," KJL-10-98, Lockheed Martin Idaho Technologies Co., Idaho Falls, Idaho, October 30, 1998.

ANL 1990, *AGHCF Operational Manual*, Section 11.0, "Waste Management," IRS-2-00-00, Argonne National Laboratory, Chicago, Illinois, September 1990.

Westinghouse, *TRUPACT-II Authorized Methods for Payload Control (TRAMPAC),* Appendix 5.2, Rev. 19a, Westinghouse TRU Solutions, LLC, Carlsbad, New Mexico, March 2002.

S. T. Khericha, R. N. Bhatt, and K. J. Liekhus, *Methodology to Predict Hydrogen Concentration in RH-TRU Waste Drums*, INEEL/EXT-02-01250, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho, February 2003.

Westinghouse, *Safety Analysis Report for the TRUPACT-II Shipping Package*, Rev. 19a, Westinghouse Electric Corporation, Waste Isolation Division, Carlsbad, New Mexico, March 2002.

J. Brandup, E. H. Immergut, and E. A. Grulke, *Polymer Handbook,* 4th ed., New York: John Wiley and Sons, 1999.

S. T. Khericha, *Estimation of Time to Build-up Hydrogen Concentration 2.2% in Sealed ILTSF Vaults After Purging* , Idaho National Engineering and Environmental Laboratory, Engineering Design File 5015, July 2004.

S. T. Khericha, *Identification of RH-TRU Waste Drums with a Potential Hydrogen Concentration of Greater Than the Lower Explosive Limit (4.4%)*, Idaho National Engineering and Environmental Laboratory, Engineering Design File 4328, November 2003.

K. J. Liekhus, correspondence with M. J. Connolly, "Hydrogen Diffusion Characteristic across Drum Gaskets," KJL-04-97, Lockheed Martin Idaho Technologies Co., Idaho Falls, Idaho, September 1997.

AGHCF Operations Manual, Section 11.0, "Waste Management," Argonne National Laboratory, IPS-2-00-00, Rev. 3, Oct. 22, 1996.